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Giant and universal magnetoelectric coupling in soft materials and concomitant ramifications for materials science and biology

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Magnetoelectric coupling—the ability of a material to magnetize upon application of an electric field and, conversely, to polarize under the action of a magnetic field—is rare and restricted to a rather small set of exotic hard crystalline materials. Intense research activity has recently ensued on materials development, fundamental scientific issues, and applications related to this phenomenon. This tantalizing property, if present in adequate strength at room temperature, can be used to pave the way for next-generation memory devices such as miniature magnetic random access memories and multiple state memory bits, sensors, energy harvesting, spintronics, among others. In this Rapid Communication, we prove the existence of an overlooked strain mediated nonlinear mechanism that can be used to *universally* induce the giant magnetoelectric effect in all (*sufficiently*) soft dielectric materials. For soft polymer foams—which, for instance, may be used in stretchable electronics—we predict room-temperature magnetoelectric coefficients that are comparable to the best known (hard) composite materials created. We also argue, based on a simple quantitative model, that magnetoreception in some biological contexts (e.g., birds) most likely utilizes this very mechanism.

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Despite their discovery as early as in the 1950's, magnetoelectric materials remained a scientific curiosity for several decades. Single-phase, intrinsically magnetoelectric materials are only weakly so, and that too at impractically low temperatures—in fact, to date, a *single-phase* material that exhibits a strong enough magnetoelectric effect for room-temperature operable technological applications has yet to be discovered [1–10]. Recent advances in fabrication methods, nanotechnology, and concurrent insights from *ab initio* calculations have spurred intense research in this field [1,2,11,12]. Indeed, several exciting applications appear to be on the horizon based on magnetoelectric materials, e.g., wireless energy transfer [4], spintronics, multiple-state memory bits [8], nonvolatile memories, among others [3]. In the case of memories, for instance, the magnetoelectric effect could in principle allow data to be written electrically and read and retrieved magnetically [2].

The scarcity and the weak magnetoelectricity in single-phase materials can be partially understood from symmetry and atomistic considerations [11,13]. Ferroelectrics must be dielectric while ferromagnetic materials are usually metallic. Additionally, ferroelectrics require broken centrosymmetry while ferromagnetics require broken time-reversal symmetry. The stringent symmetry constraint, and the contradictory electronic structure of the typical ferromagnets and ferroelectrics, allow very few materials to support the magnetoelectric effect [2,8,14]. An alternative and promising approach to engineer magnetoelectricity is through the development of composite materials that indirectly connect electrical and magnetic degrees of freedom through a third order parameter (e.g., mechanical deformation). A typical such scheme is to create composites of piezoelectric (usually ferroelectric) and magnetostrictive materials [12,15–18]. In such a composite, an applied magnetic field will cause the magnetostrictive component to deform. The mechanical strain is transferred to the adjoining piezoelectric phase. The strain in the piezoelectric phase then produces an electric field. Notably, while neither of the constituent materials are magnetoelectric, overall, they function as such. For large magnetoelectric coupling, the magnetostrictive and piezoelectric coefficients must be quite high. The latter, despite many advances, is not simple to achieve. Furthermore, brittle ceramics are the most commonly used constituents with their accompanying disadvantages of fabrication, expense, scarcity, and small deformation. Soft magnetoelectric materials, if available, would yield orders of magnitude higher deformation than currently possible. Further, they will also find application in stretchable electronics, nonplanar configurations [19], memory devices, and have the additional advantage of ease of manufacturability and cost effectiveness. We note here that fairly extensive work on soft multifunctional materials exists in the purely electrostatic context—see, for example, Refs. [20,21] and references therein.

In this Rapid Communication we report a mechanism that does not require creating complex composites and can be *universally* employed for all *soft* dielectric materials to create a giant magnetoelectric effect. In addition, aided by the presence of externally deposited magnetic dipoles in soft polymers (*that do not have to be piezoelectric*), the magnetoelectric coefficient may be increased to values that are comparable to the best known composites. Our suggested mechanism, although also strain mediated, neither requires the use of composites nor piezoelectric or magnetostrictive materials.

Before presenting a quantitative calculation, the central physical concept is explained via Fig. 1. We consider a soft homogeneous dielectric thin film as sketched in Fig. 1(a).

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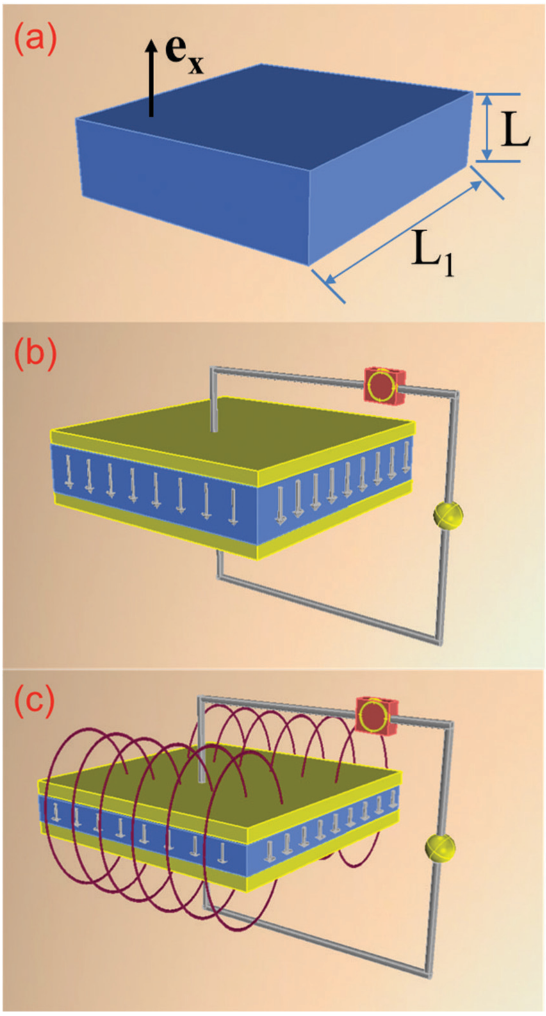

FIG. 1. (Color online) The schematic and the central concept. (a) Visualize a homogeneous dielectric thin film that is neither magnetostrictive nor piezoelectric. The material has a magnetic permeability different from that of vacuum (i.e., $\mu_r > 1$). (b) The thin film is placed under bias and the film is uniformly polarized. Due to the electric Maxwell stress, the thin film is compressed. (c) The compressed thin film (with the preexisting electric field) is now placed in a magnetic field. The magnetic Maxwell stress now also deforms the thin film which alters the preexisting electric field. The change in electric field can be measured as current.

The materials are assumed to be elastically nonlinear, and electrostatically and magnetostatically linear, with constitutive relations given by

$$\tilde{\boldsymbol{\Sigma}}_{\text{mech}} = \frac{\partial W_{\text{elst}}(\mathbf{F})}{\partial \mathbf{F}}, \quad \mathbf{e} = \frac{\mathbf{p}}{\epsilon_0(\epsilon_r - 1)}, \quad \mathbf{h} = \frac{\mathbf{m}}{\mu_r - 1}, \tag{1}$$

where $\mathbf{F}$, $\mathbf{p}$, and $\mathbf{m}$ are the deformation gradient, polarization, and magnetization, respectively; $\tilde{\boldsymbol{\Sigma}}_{\text{mech}}$, $\mathbf{e}$, and $\mathbf{h}$ are the mechanical Piola-Kirchhoff stress, electric field, and magnetic field, respectively; $W_{\text{elst}} = W_{\text{elst}}(\mathbf{F})$ is the elastic energy density function, and μ_r (respectively ϵ_r) is the relative magnetic permeability (respectively electric permittivity) of the elastomer. Since the material is homogeneous, nonpiezoelectric, and nonmagnetostrictive, there should be no intrinsic coupling between polarization and magnetization, i.e., no intrinsic magnetoelectricity. This is well evident from the constitutive relations in (1). We apply a uniform external electric field to this material. This may alternatively be also achieved by embedding charge in the soft material, as done experimentally in electret materials [22]. The electrical Maxwell stress will tend to compress the material and the film will polarize [Fig. 1(b)]. Further, we apply an external magnetic field that will also exert a *magnetic* Maxwell stress on the film and induce an additional change in the thickness of the film. As shown in Fig. 1(c), this ensuing change in thickness will in turn change the state of the preexisting electric field and hence polarization. The converse effect can be similarly explained if there are external magnetic dipoles embedded in the film or a preexisting applied magnetic field exists.

Several observations may be made: (1) The aforementioned mechanism is *universal* for all dielectric films. (2) The aforementioned mechanism does not require the materials to be magnetostrictive or piezoelectric and instead relies on the universal (and nonlinear) Maxwell stress effect. (3) How strong is this effect? The material must be *soft*. From a practical viewpoint, the magnetic field ($\mathbf{H}$ field) can reach up to the order of 10^4 Oe (i.e., the magnetic flux $\mathbf{B}$ is 1 T) and the dielectric strength of typical materials is at the order 10^6–10^8 V/m. Therefore, the upper bound on the Maxwell stress ($\sim\epsilon_0|\mathbf{E}|^2 + \mu_0|\mathbf{H}|^2$) is roughly 10^5 Pa, corresponding to 10% strain for a material of Young's modulus of 1 MPa. Therefore, the magnetoelectric coupling induced by the Maxwell stress is only significant for soft polymer-based materials that require a geometrically nonlinear theory of elasticity. (4) The magnetic permeability of the material must differ from that of the vacuum. As an example, this condition may be ensured and improved for all soft materials by simply embedding a minute amount of magnetic dipoles (e.g., soft magnetites). We note here that there does exist a magnetic polymer [23] which may be quite useful in the context of our proposed mechanism (albeit, by no means, necessary). (5) The strength of the coupling may be tuned by the applied voltage V or the magnetic field (for the converse effect).

We now present a quantitative explanation and the geometrically nonlinear theory underpinning the central mechanism that was heuristically explained above. The thermodynamic state of the film in Fig. 1 is described by the deformation, polarization, and magnetization (per unit volume in the reference configuration) $[\boldsymbol{\chi}(\mathbf{X}), \tilde{\mathbf{P}}(\mathbf{X}), \tilde{\mathbf{M}}(\mathbf{X})]$, where $\mathbf{X} = (X, Y, Z)$ are the Lagrange coordinates of the material points. Since the central idea is related to the nonlinear deformation state, the distinction between the reference and deformed configuration must be carefully maintained. For simplicity (and without impacting the central physics), we suppose that the conducting electrodes maintain constant electrostatic potentials on the top and bottom faces and are mechanically trivial (i.e., zero stiffness and zero thickness), and the film is in a uniform external magnetic field $\mathbf{h}^e$ that permeates to the entire space. Since the film is thin (the thickness L is much smaller than the width L_1 in the other two directions), we can safely neglect the electric field in the ambient medium (i.e., the fringe electric field). In addition, parallel to the classic theory for axially loaded members, we assume that the deformation $\mathbf{x} = \boldsymbol{\chi}(\mathbf{X})$

to be of the form

$$x = X + u(X), \tag{2}$$

$y = Y(1+\epsilon_y(X))$, $z = Z(1+\epsilon_z(X))$, and that the polarization $\tilde{\mathbf{P}} = \tilde{P}(X)\mathbf{e}_x$ is along the $\mathbf{e}_x$ direction and depends only on X, where $\mathbf{x} = (x,y,z)$ are the Euler coordinates and $u,\epsilon_y,\epsilon_z : (0,L) \to \mathbb{R}$ are scalar functions describing the deformed state of the film. Denote the deformation gradient by $\mathbf{F} = \text{Grad}\,\boldsymbol{\chi}$, the *stretches* in the X (respectively Y,Z) direction by $\lambda_1 = 1 + \frac{\partial u}{\partial X}$ (respectively $\lambda_2 = \frac{\partial y}{\partial Y} = 1+\epsilon_y, \lambda_3 = \frac{\partial z}{\partial Z} = 1 + \epsilon_z$), and $J = \det \mathbf{F} = \lambda_1\lambda_2\lambda_3$ the Jacobian measuring the change of volume. In the current configuration the polarization $\mathbf{p}$ and magnetization $\mathbf{m}$ on the film are given by

$$\mathbf{p} = \tilde{\mathbf{P}}/J, \quad \mathbf{m} = \tilde{\mathbf{M}}/J. \tag{3}$$

Without loss of generality we assume that $x(0) = 0$ and $x(L) = l$. Clearly, the electric field $-\text{grad}\,\xi$ and the magnetic field $-\text{grad}\,\zeta$ in the *current configuration* must satisfy the Maxwell equations

$$\text{div}(-\epsilon_0\,\text{grad}\,\xi + \mathbf{p}) = 0, \quad \text{div}(-\text{grad}\,\zeta + \mathbf{m}) = 0, \tag{4}$$

for the boundary conditions $\xi(x = l) = V$, $\xi(x = 0) = 0$, and $-\text{grad}\,\zeta \to \mathbf{h}^e$ as $|\mathbf{x}| \to +\infty$. We first observe that, as implied by the constitutive laws (1), the stored or internal energy density function of the material is of the following form [24]:

$$\Psi(\mathbf{F},\tilde{\mathbf{P}},\tilde{\mathbf{M}}) = W_{\text{elst}}(\mathbf{F}) + \frac{|\tilde{\mathbf{P}}|^2}{2\epsilon_0(\epsilon_r - 1)J} + \frac{\mu_0|\tilde{\mathbf{M}}|^2}{2(\mu_r - 1)J}. \tag{5}$$

For ease of exposition, we assume that the material is elastically isotropic, a reasonable assumption for soft polymers. Our central idea is not sensitive to the choice of particular elasticity model and here we employ the Neo-Hookean hyperelastic model with $W_{\text{elst}}(\mathbf{F})$ given by (μ is the shear modulus, κ is the bulk modulus)

$$W_{\text{elst}} = \frac{\mu}{2}\left[J^{-2/3}\left(\lambda_1^2 + \lambda_2^2 + \lambda_3^2\right) - 3\right] + \frac{\kappa}{2}(J-1)^2. \tag{6}$$

Taking the applied voltage and external magnetic field into account, we identify the total free energy of the system as [24,25]

$$\begin{aligned}\frac{1}{L_1^2}&F[\boldsymbol{\chi},\tilde{\mathbf{P}},\tilde{\mathbf{M}}]\\ &= \int_0^L \Psi(\mathbf{F},\tilde{\mathbf{P}},\tilde{\mathbf{M}})dX\\ &\quad + \frac{\epsilon_0}{2}\int_0^l \lambda_2\lambda_3|\text{grad}\,\xi|^2 dx + V\lambda_2\lambda_3(-\epsilon_0\,\text{grad}\,\xi + \mathbf{p})|_{x=l}\\ &\quad + \int_0^l \lambda_2\lambda_3\frac{\mu_0}{2}|\text{grad}\,\zeta^{\text{self}}|^2 - \int_0^l \lambda_2\lambda_3\mu_0\mathbf{h}^e\cdot\mathbf{m}dx,\end{aligned} \tag{7}$$

where $-\text{grad}\,\zeta^{\text{self}} := -\text{grad}\,\zeta - \mathbf{h}^e$ is the self-magnetic field induced by the magnetized film alone. By the principle of minimum free energy, we conclude that the equilibrium state of the film is dictated by the variational problem

$$\min_{(\boldsymbol{\chi},\tilde{\mathbf{P}},\tilde{\mathbf{M}})} F[\boldsymbol{\chi},\tilde{\mathbf{P}},\tilde{\mathbf{M}}]. \tag{8}$$

By a standard first variation calculation we immediately find that in equilibrium the polarization $\mathbf{p}$ and magnetization $\mathbf{m}$ necessarily satisfy that $\mathbf{p} = -\epsilon_0(\epsilon_r - 1)\text{grad}\,\xi$ and $\mathbf{m} = (\mu_r - 1)(-\text{grad}\,\zeta^{\text{self}} + \mathbf{h}^e)$, which are precisely the second and third constitutive relations in (1). Inserting these constitutive relations (1) into the Maxwell's equations (4) we find that the electric field $-\text{grad}\,\xi$ and polarization for $x \in (0,l)$ are given by

$$-\text{grad}\,\xi = -\frac{V}{l}\mathbf{e}_x, \quad \mathbf{p} = -\epsilon_0(\epsilon_r - 1)\frac{V}{l}\mathbf{e}_x. \tag{9}$$

Also, if the external magnetic field $\mathbf{h}^e = h_x^e\mathbf{e}_x$, the self-magnetic field $-\text{grad}\,\zeta^{\text{self}}$ and magnetization for $x \in (0,l)$ are given by

$$-\text{grad}\,\zeta^{\text{self}} = -\frac{\mu_r - 1}{\mu_r}h_x^e\mathbf{e}_x, \quad \mathbf{m} = \frac{\mu_r - 1}{\mu_r}h_x^e\mathbf{e}_x, \tag{10}$$

whereas if the external magnetic field $\mathbf{h}^e = h_y^e\mathbf{e}_y$, the self-magnetic field $-\text{grad}\,\zeta^{\text{self}}$ and magnetization is given by

$$-\text{grad}\,\zeta^{\text{self}} = 0, \quad \mathbf{m} = (\mu_r - 1)h_y^e\mathbf{e}_y. \tag{11}$$

Therefore, by (3), (5), (7), (9), (10), (11), and straightforward algebraic calculations, we rewrite the free energy in terms of $\lambda_1 = l/L, J$ as

$$\frac{F(\lambda_1,J)}{\mu L L_1^2} = \hat{W}_{\text{elst}}(\lambda_1,J) - \frac{J}{2\lambda_1^2}\hat{f}^{\text{elct}} - \frac{J}{2}\hat{f}^{\text{mgf}}, \tag{12}$$

where the following dimensionless quantities are introduced for clarity:

$$\begin{aligned}\hat{W}_{\text{elst}}(\lambda_1,J) &= \frac{1}{2}\left[J^{-2/3}\left(\lambda_1^2 + \frac{2J}{\lambda_1}\right) - 3\right] + \frac{\hat{\kappa}}{2}(J-1)^2,\\ \hat{\kappa} &= \frac{\kappa}{\mu}, \quad \hat{f}^{\text{elct}} = \frac{\epsilon_r\epsilon_0 V^2}{\mu L^2},\\ \hat{f}^{\text{mgf}} &= \frac{\mu_0}{\mu}\left[\frac{\mu_r - 1}{\mu_r}\left(h_x^e\right)^2 + (\mu_r - 1)\left(h_y^e\right)^2\right].\end{aligned} \tag{13}$$

The equilibrium (λ_1,J) shall be such that $F(\lambda_1,J)$ is minimized, and hence necessarily satisfy $\frac{\partial F}{\partial\lambda_1} = \frac{\partial F}{\partial J} = 0$, i.e.,

$$\begin{aligned}J^{-2/3}\lambda_1 - J^{1/3}\lambda_1^{-2} + J\lambda_1^{-3}\hat{f}^{\text{elct}} &= 0,\\ -\tfrac{1}{3}\left(J^{-5/3}\lambda_1^2 - J^{-2/3}\lambda_1^{-1}\right) + \hat{\kappa}(J-1)&\\ -\tfrac{1}{2}\lambda_1^{-2}\hat{f}^{\text{elct}} - \tfrac{1}{2}\hat{f}^{\text{mgf}} &= 0.\end{aligned} \tag{14}$$

From (14) we observe that the stretching λ_1 and hence the actual electric field along the $\mathbf{e}_x$ direction $E = -\xi_{,x} = -\frac{V}{L\lambda_1}$ depend on the magnitude and direction of the external magnetic field $\mathbf{h}^e$. From this viewpoint, the magnetoelectric coupling is *effectively* created from the Maxwell stress and geometric nonlinearity. The change of electric field at the presence of an external magnetic field is appropriate for evaluating the strength of this magnetoelectric coupling, which is given by

$$E = -\frac{V}{L}\left(\lambda_1^{-1}\big|_{\mathbf{h}^e\neq 0} - \lambda_1^{-1}\big|_{\mathbf{h}^e=0}\right). \tag{15}$$

Immediately, we also find the change of polarization $p = \epsilon_0(\epsilon_r - 1)E$.

We can numerically solve (14) and determine the change of electric field E induced by the external magnetic field;

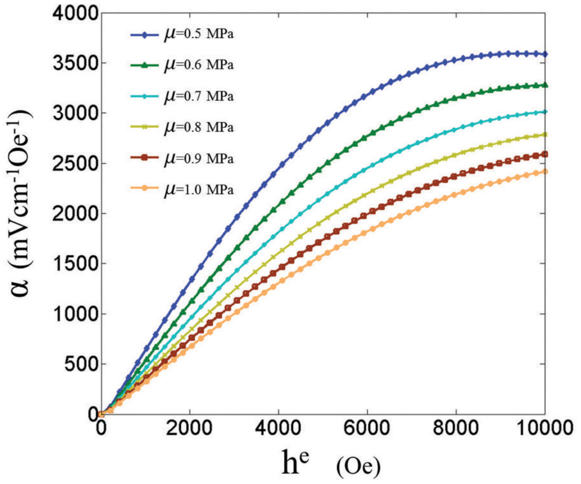

FIG. 2. (Color online) Magnetoelectric coupling constants of soft films with $\epsilon_r = 20$, $\hat{\kappa} = 14/3$, and $\tilde{E}_0 = 10^7$ V/m.

the dependence of the *magnetoelectric coupling constant* $\alpha := E/h^e_y$ on the in-plane magnetic field strength h^e_y is shown in Fig. 2 for a fixed nominal electric field $\tilde{E}_0 := V/L = 10^7$ V/m ($\epsilon_0|\tilde{E}_0|^2 = 885.4$ Pa). The material properties are chosen as $\epsilon_r = 20, \mu_r = 5, \hat{\kappa} = 14/3$ (corresponding to a Poisson ratio 0.4), and $\mu = 1, 0.9, 0.8, 0.7, 0.6,$ and 0.5 MPa. From Fig. 2 we observe that the magnetoelectric coupling constant depends linearly on h^e_y initially and then deviates for larger h^e_y; the dependence of the magnetoelectric coupling constant on the external magnetic field (and nominal electric field) is qualitatively different from the conventional magnetoelectric composites or multiferroic crystals. The magnetoelectric coefficients depicted in Fig. 2 compare well with some well-known composites, e.g., at a field of 300 Oe: A textured NCZF-PZT-PZN-NCZF trilayer system has an α of 900 mV/cm Oe, which drops to 200 at a field strength of 500 Oe [9]. In general, a direct comparison with some other claimed high magnetoelectric composites (e.g., Tefernol-D/PZT [26]) is difficult since our results are essentially frequency independent, are capable of orders of magnitude larger deformations, and finally may yield higher polarization for the same magnetoelectric coefficient. Additionally, several composite configurations rely on the proximity of the constituent ferroelectric materials to phase transformation conditions.

Figure 3 shows the dependence of p on the direction of the angle θ for a fixed nominal electric field $\tilde{E}_0 = 10^7$ V/m and external magnetic field $|\mathbf{h}^e| = 10^6$ A/m; the shear modulus $\mu = 0.5$ MPa and $\hat{\kappa} = 14/3, 10, 20, 30, 40$.

We remark here that the mechanism pointed out by us is not at odds with the ongoing composite development efforts and indeed may be combined with those to leverage this nonlinear effect to further improve materials design.

From Fig. 2 we observe that the induced electric field by the external magnetic field is roughly proportional to h^e and the slope of the curves increases as the shear modulus μ or bulk modulus κ decrease. To calculate the slope explicitly, we assume small strain $\lambda := \lambda_1 - 1 \sim J := J - 1 \sim \eta \ll 1$, expand and truncate the free energy (12) up to the order of η^2, neglect an immaterial (λ_1, J)-independent constant, and, after imposing the necessary conditions $\partial F/\partial\lambda = \partial F/\partial J = 0$ for the equilibrium (λ, J), obtain

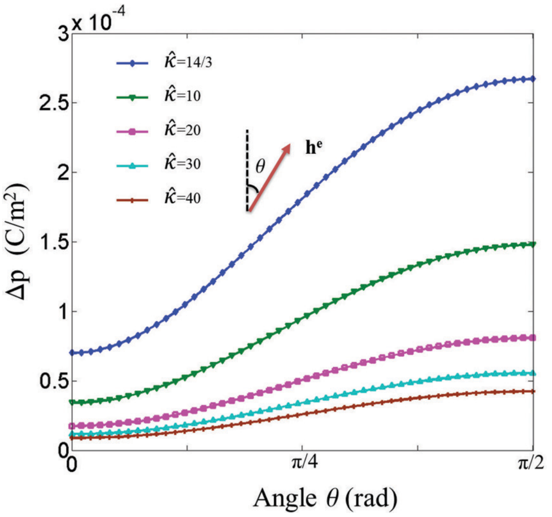

FIG. 3. (Color online) The change of polarization vs the direction of applied magnetic field. The magnitude of the magnetic field $|\mathbf{h}^e| = 10^4$ Oe, applied nominal electric field $\tilde{E}^0 = 10^7$ V/m, and shear modulus $\mu = 0.5$ MPa.

$$\begin{aligned} &3(1 - \hat{f}^{\text{elct}})\lambda - (1 - \hat{f}^{\text{elct}})J + \hat{f}^{\text{elct}} = 0, \\ -&(1 - \hat{f}^{\text{elct}})\lambda + \left(\tfrac{1}{3} + \hat{\kappa}\right)J - \tfrac{1}{2}\hat{f}^{\text{elct}} - \tfrac{1}{2}\hat{f}^{\text{mgf}} = 0. \end{aligned} \tag{16}$$

Solving the above equations we finally obtain the change of electric field (15):

$$E = \tilde{E}_0 \frac{\hat{f}^{\text{mgf}}}{2(3\hat{\kappa} + \hat{f}^{\text{elct}})}. \tag{17}$$

While the implications for materials science are obvious from the preceding discussions, we also comment on the possible relevance of our suggested mechanism in biology. Certain anaerobic bacteria, flighted birds, among many other life forms, possess magnetoreception—see, e.g., Refs. [27–30]. The central mechanisms underpinning biological magnetoreception, however, are still under debate. For instance, in the case of avian magnetoreception [30], no magnetoelectric organ or material constituent has been found. While small iron-oxide "magnetite" crystals with a relative magnetic permeability μ_r at the order of 10^2–10^5 are found in magnetoreceptive cells, the mechanism that will allow magnetic signals to be interpreted electrically has not been elucidated. Here we show that our proposed mechanism provides an experimentally testable explanation. To make quantitative estimates, we consider a lipid bilayer with embedded magnetite particles. A typical cell membrane has a nominal electric field at the order of 4.0×10^6 V/m across the membrane due to an ion imbalance of actively gated transportation whereas neurons can sense an electric field change as low as 0.1 V/m [31], which is 0.025 ppm of the nominal electric field. Since the earth magnetic field is about 0.6 Oe on its surface, the change of electric field due to the change of direction of the magnetic field, given by (17), is estimated to be $E \approx \tilde{E}_0 \frac{5\times10^{-4}\mu_r}{\kappa}$. Therefore, if $\mu_r/\kappa \geqslant 5 \times 10^{-5}$ Pa^{-1}, the change of electric field will be sensible to neurons. In reality, we however anticipate that the presented model oversimplifies the elastic and magnetic behaviors of the lipid bilayer membrane, ignoring the out-of-plane anisotropy of the membrane and replacing the magnetic effect of ferrite particles by an effective permeability μ_r. A more detailed derivation of the resulting electric field change on account of these effects is rather tedious and will be presented elsewhere, however, we have verified that our estimates are reasonable, thus ensuring the plausibility of our argument.

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